

PATENT ABSTRACTS OF JAPAN

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(54) SCINTILATOR AND RADIATION DETECTOR WITH IT

(57)Abstract:

PROBLEM TO BE SOLVED: To increase luminous efficiency and to shorten the decay time by forming a scintillator with a crystal emitting fluorescence with the prescribed decay time in response to the incidence of radiation, and setting the decay time of the fluorescence component to a prescribed value or below at the room temperature.

SOLUTION: This scintillator is made of a crystal having the composition expressed by the general formula $\text{CsBr}_{1-x}\text{I}_x$, where (x) is the ratio (mole fraction) of the number of I atoms against the total number (mole number) of Br atoms and I atoms, and $0.01 \leq x \leq 0.1$. If $x < 0.01$, fluorescence intensity becomes weak. If $x > 0.1$, no mixed crystal is generated, I is deposited, and a lattice defect is increased, thus (x) is preferably set to $0.05 \leq x \leq 0.08$. The decay time of the scintillator at the room temperature in response to the incidence of radiation is 2 ns or below, preferably 1.5 ns or below. Fluorescence including the fluorescence component with the very short decay time is emitted from the crystal of the composition. The resolution of a detector using the scintillator having a shorter decay time can be improved, and the decay time is preferably set to 100 ps or above.

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CLAIMS

[Claim(s)]

[Claim 1] the scintillator which emits the fluorescence containing the fluorescence component which has the predetermined damping time according to the incidence of a radiation -- it is -- this scintillator -- following general formula: $\text{CsBr}_{1-x}\text{I}_x$ [-- x are the number of 0.01-0.1 among a formula.] The scintillator to which it consists of a crystal which comes out and has the presentation expressed, and the damping time of said fluorescence component is characterized by what is been 2 or less ns at a room temperature.

[Claim 2] The scintillator according to claim 1 to which said fluorescence contains two or more fluorescence components which have the predetermined damping time, and the damping time of at least one component in this fluorescence component is characterized by being 2 or less ns at a room temperature.

[Claim 3] said crystal -- following general formula: $\text{CsBr}_{1-x}\text{I}_x$ [-- x are the number of 0.01-0.1 among a formula.] The scintillator according to claim 1 or 2 characterized by making the mixed powder of CsBr and CsI which were mixed so that it might come out and might become the presentation expressed crystalize with a perpendicular Bridgman method.

[Claim 4] the scintillator which emits the fluorescence containing the fluorescence component which has the predetermined damping time according to the incidence of a radiation -- it is -- this scintillator -- following general formula: $\text{CsBr}_{1-x}\text{I}_x$ [-- x are the number of 0.01-0.1 among a formula.] The radiation detector characterized by having the fluorescence detector which detects the fluorescence which consists of a crystal which comes out and has the presentation expressed and, by which the damping time of said fluorescence component was emitted at the room temperature from the scintillator which is 2 or less ns, and said scintillator.

DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention] This invention relates to the scintillator which emits fluorescence according to the incidence of a radiation, and the radiation detector which used it for the list.

[0002]

[Description of the Prior Art] Conventionally, in inorganic scintillator, the damping time (it is also called decay time and the life time of fluorescence) is comparatively short, and

the CsI (cesium iodide) scintillator has been widely used for detection and medical equipment of a high energy particle as a thing which a measurement rate can improve. However, if it is in the conventional CsI scintillator Luminous efficiency is as small as 4 - 5%. Moreover, the damping time of a quick fluorescence component is as short never enough as 10 - 16ns. there was no ** (it Ruan(s) S. -- Kubota and J. --) M. Itoh and S.Hashimoto, S. Sakuragi"A New Type of Luminescence Mechanism in Large Band-Gap Insulators: Proposal for Fast Scintillation Materials" Nuclear Instruments and Methods in Physics Research A289 (1990) p.253-260.

[0003] On the other hand, to CsI, Rb (rubidium), K (potassium), Br (bromine), The scintillator which consists of a crystal which doped Cl (chlorine) etc. as an impurity is examined. By doping little Br to CsI It is indicated that an attenuation rate improves (A.). [Gektin, N.Shiran, V.Shlyahurov,] [A.Belsky"Development ofFast Scintillators on] the Basis of CsI Doped with Homological Impurities" Proc.Int.Conf.on Inorganic Scintillators and Their Applications, SCINT95 (1996) Delft University Press, The Netherlands, p.415-418. However, even if it was the case where little Br was doped to CsI, the reduction in the damping time was very slight, it was not yet enough, and when Br concentration became high, the crystal became unstable, and on the other hand, if decomposition occurred, it is recognized conventionally.

[0004]

[Problem(s) to be Solved by the Invention] This invention is made in view of the technical problem which the above-mentioned conventional technique has, and aims at providing with a radiation detector with high resolution the scintillator which emits the fluorescence in which luminous efficiency contains a fluorescence component with the very short damping time in a room temperature highly, and a list.

[0005]

[Means for Solving the Problem] this invention persons came to complete a header and this invention for the scintillator which emits the fluorescence in which luminous efficiency contains a fluorescence component with the very short damping time highly being obtained, when little I is doped to CsBr, as a result of repeating research wholeheartedly that the above-mentioned purpose should be attained.

[0006] namely, the scintillator which emits the fluorescence in which the scintillator of this invention contains the fluorescence component which has the predetermined damping time according to the incidence of a radiation -- it is -- this scintillator -- following general formula: $\text{CsBr}_{1-x}\text{I}_x$ [-- x are the number of 0.01-0.1 among a formula.] It consists of a crystal which comes out and has the presentation expressed, and the damping time of said fluorescence component is characterized by what is been 2 or less ns at a room temperature.

[0007] moreover, the scintillator which emits the fluorescence in which the radiation detector of this invention contains the fluorescence component which has the predetermined damping time according to the incidence of the (a) radiation -- it is -- this scintillator -- following general formula: $\text{CsBr}_{1-x}\text{I}_x$ [-- x are the number of 0.01-0.1 among a formula.] It is characterized by having the fluorescence detector which detects the fluorescence which consists of a crystal which comes out and has the presentation expressed and, by which the damping time of said fluorescence component was emitted at the room temperature from the scintillator which is 2 or less ns, and the (b) aforementioned scintillator.

[0008]

[Embodiment of the Invention] Hereafter, the suitable operation gestalt of this invention is explained to a detail, referring to a drawing. In addition, it considers as the same or ***** which gives the same sign to a considerable part among a drawing.

[0009] First, the scintillator of this invention is explained.

[0010] The scintillator of this invention consists of a crystal which has the presentation expressed with following general formula: $\text{CsBr}_{1-x}\text{I}_x$. x in an upper type is the ratio (mole fraction) of the number of I atoms to the total number (the number of mols) of Br (bromine) atom under crystal, and I (iodine) atom, and when using the mixture of CsBr (cesium bromide) and CsI (cesium iodide) as a raw material, it is equivalent to the mole fraction of CsI in a raw material.

[0011] In this invention, it is required for Above x to be the number of or more 0.01 0.1 or less range. Fluorescence intensity becomes [x] weak less than by 0.01, if another side and x exceed 0.1, mixed crystal will not be made, but I deposits, and a lattice defect increases. Moreover, as for Above x , it is desirable that it is the number of or more 0.05 0.08 or less range. The number of I₂ decreases under at the above-mentioned minimum, x is in the inclination for fluorescence intensity to become weak, if another side and x exceed the above-mentioned upper limit, the lattice defect by the deposit of I will increase, and it is in the inclination for the component of the early damping time to decrease.

[0012] In addition, according to the incidence of a radiation, as for the scintillator of above-mentioned this invention, the damping time in a room temperature emits preferably the fluorescence which comes out 1.5 or less ns and contains a certain fluorescence component 2 or less ns. Thus, that the fluorescence in which the damping time in a room temperature contains the fluorescence component with the very short damping time of 2 or less ns is emitted from the crystal of the above-mentioned presentation is exactly the fact which has not been recognized at all conventionally but was found out for the first time by this invention person. And since the fluorescence emitted by the scintillator of this invention contains a fluorescence component with the very short damping time in a room temperature as mentioned above, a radiation detector with very high resolution is obtained by using the component. On the other hand, since the damping time is in the inclination exceeding the resolving power of the fluorescence detector used combining a scintillator in less than 100 ps, as for the above-mentioned damping time, it is desirable [improvement in the resolving power of the radiation detector using the scintillator is attained so that the damping time is short, but] that they are 100 or more ps. In addition, 300K (27 degrees C) are called "room temperature" here.

[0013] Moreover, the fluorescence emitted by the scintillator of this invention usually contains two or more attenuation components (fluorescence component) based on the luminescence mechanism, and the damping time of at least one component in those attenuation components (the so-called early component (fast component)) is 2 or less ns at a room temperature in that case. Thus, since the fluorescence emitted by the scintillator of this invention contains at least one fluorescence component with the very short damping time in a room temperature, a radiation detector with very high resolution is obtained by using the component.

[0014] In addition, the "damping time" here is the value which analyzed and asked for the

decay curve of the fluorescence emitted by the scintillator according to the following approaches. That is, first, when fluorescence consists of one component, the decay curve is expressed with the following exponential-function: $F(t) = k \cdot e^{-t/\tau}$ [the function of fluorescence intensity [as opposed to / as opposed to / in t / time amount / time amount in $F(t)$] and k show the initial value ($k=F(0)$) of fluorescence intensity among a formula, and τ shows the damping time, respectively]. And the damping time is time amount until fluorescence intensity declines to $1/e$ of initial value, and is a value with which the following related : $F(\tau)/F(0) = 1/e$ are filled.

[0015] Moreover, when fluorescence consists of two components, as it is shown in drawing 1 , the decay curve is the sum of two exponential functions, i.e., the following

functions. :

The function of the inside of [type, and fluorescence intensity [as opposed to / as opposed to / in t / time amount / time amount in $F(t)$], the function of fluorescence intensity [as opposed to the time amount about the 1st component in $f_1(t)$], The function of the fluorescence intensity to the time amount about the 2nd component and k_1 $f_2(t)$ The initial value of the fluorescence intensity about the 1st component ($k_1=f_1(0)$), $k_2=f_2(0)$ -- τ_1 -- one -- the -- one -- a component -- about -- the damping time -- τ_2 -- two -- the -- two -- a component -- about -- the damping time -- respectively -- being shown --] -- expressing -- having . And the damping time about the 1st and 2nd components is time amount until the fluorescence intensity about the 1st and 2nd components declines to $1/e$ of initial value, respectively, and is a value with which the following relation: $f_1(\tau_1)/f_1(0) = 1/e$ $f_2(\tau_2)/f_2(0) = 1/e$ is filled.

[0016] Furthermore, when fluorescence consists of three or more components, similarly, the decay curve is expressed with the sum of the exponential function according to the number of components, and the damping time about each component is found as time amount until the fluorescence intensity about each component declines to $1/e$ of initial value.

[0017] Therefore, while approximating measurement data (attenuation data of fluorescence) using the least square method, it asks for the exponential function of the number according to the number of components, and the damping time about each component is found based on the obtained exponential function. And when fluorescence consists of two or more components, a component with the acquired shorter damping time is equivalent to a earlier component. In addition, such an analysis approach is an approach currently generally used as an approach of evaluating the property of a scintillator.

[0018] As a radiation (excitation light) by which incidence is carried out to the scintillator of this invention, when absorbed by the scintillator, a gamma ray, alpha rays, beta rays, an X-ray, and ultraviolet rays are mentioned that what is necessary is just what forms an excitation state during the crystal. Although the fluorescence intensity about each component changes according to this radiation (a class, photon energy), the above-mentioned damping time is not influenced of the target radiation. Moreover, although the peak wavelength of the fluorescence emitted from the scintillator of this invention changes according to the presentation, it is 287nm in general.

[0019] Furthermore, the consistency of the scintillator of this invention is comparatively as high as 4.44 g/cm³, and, so, the stopping power to radiations, such as a gamma ray, as a result its absorption coefficient are in general high. Moreover, the relative luminescence reinforcement (luminous efficiency) when making luminescence reinforcement of NaI (Tl) into 100% is 13.9 - 17.5%, and the scintillator of this invention has luminous efficiency higher than the conventional CsI scintillator.

[0020] Although especially the method of manufacturing the scintillator of above-mentioned this invention is not restricted, the so-called perpendicular Bridgman method which can compound a large-sized crystal is desirable. A perpendicular Bridgman method is an approach of dropping slowly the longwise crucible into which the melt of a sample was put in a vertical mold furnace (crystal growth furnace) with a predetermined temperature gradient, making it solidifying from the lower limit of the melt in a crucible, and obtaining a crystal. The following approaches are especially specifically desirable.

[0021] (i) -- first -- following general formula: CsBr_{1-x}I_x [-- x are the number of 0.01-0.1 among a formula.] CsBr and CsI are mixed and mixed powder (sample) is obtained so that it may come out and may become the presentation expressed.

[0022] (ii) Next, after putting in the above-mentioned mixed powder into a longwise crucible (for example, quartz container), the end crater which makes the inside of a crucible the vacua (degree of vacuum: 10⁻³ - 10⁻⁵Torr) is sealed.

[0023] (iii) After that, the above-mentioned crucible is arranged on the elevated-temperature band of a vertical mold furnace with a predetermined temperature gradient (elevated-temperature band: 640-670 degrees C, low-temperature band: 420-450 degree C), and the above-mentioned mixed powder is changed into a melt condition.

[0024] (iv) Then, the above-mentioned crucible is slowly dropped on a low-temperature band from the above-mentioned elevated-temperature band so that a temperature fall rate may serve as 1.2-4.6 degrees C/hr, and the above-mentioned melt is made to crystallize from the lower limit. The crystal which constitutes the scintillator of above-mentioned this invention is obtained efficiently and certainly by it.

[0025] Next, the radiation detector of this invention is explained. One suitable operation gestalt of the radiation detector of this invention is shown in drawing 2. The radiation detector shown in this drawing is equipped with the scintillator 10 of above-mentioned this invention, and the photomultiplier tube 20 as a fluorescence detector. The scintillator 10 is arranged in the tubed container 11, and, specifically, the antireflection film 14 is formed in radiation input screens 13 other than fluorescence output screen 12 of the tubed container 11. Moreover, the photomultiplier tube 20 is equipped with the hermetic container 22 which has the fluorescence input screen 21, the photoelectric surface 23 is formed inside the fluorescence input screen 21, and the convergence electrode 24, the dynode 25, and the anode 26 are further arranged in the hermetic container 22. And it is stuck to the fluorescence output screen 12 and the fluorescence input screen 21 by coupling material, such as a silicone oil.

[0026] When incidence of the radiation 31 is carried out to the scintillator 10 of such a radiation detector from a radiation source 30 and it collides with the atom in a scintillator 10, an excitation state is formed in primary and fluorescence 32 is ****(ed). Subsequently, the electron within the photoelectric surface 23 is excited by the fluorescence which penetrated the fluorescence output screen 12 and the fluorescence input screen 21, and a photoelectron 33 is emitted into a vacuum. And after it converges a

photoelectron 33 with the convergence electrode 24 and multiplication (secondary electron multiplication) is carried out by the dynode 25, a secondary electron group (charge) is taken out from an anode 26.

[0027] Thus, the fluorescence of the amount (reinforcement) proportional to the energy of a radiation occurs in a scintillator 10, and since the charge proportional to the amount of fluorescence (fluorescence intensity) is outputted by the photomultiplier tube 20, the output pulse wave height of the photomultiplier tube 20 is proportional to the energy of a radiation as a result. Therefore, the energy distribution and dosage of a radiation can be measured by measuring the peak value and the counting rate of an output pulse. In addition, since the fluorescence emitted by the scintillator of this invention contains the fluorescence component with the very short damping time of 2 or less ns in a room temperature as mentioned above, improvement in the resolution of a radiation detector of it is attained by using the component.

[0028] As mentioned above, although the suitable operation gestalt of the radiation detector of this invention was explained, the radiation detector of this invention is not limited to the above-mentioned operation gestalt. For example, a fluorescence detector may be a photodiode etc. although the photomultiplier tube is used as a fluorescence detector with the above-mentioned operation gestalt. Moreover, although the photomultiplier of the Rhine focus mold is used with the above-mentioned operation gestalt, you may be the other mold (for example, a box mold, a circular cage mold, a Venetian blind mold, a fine mesh mold, a micro-channel-plate mold).

[0029]

[Example] Hereafter, although this invention is more concretely explained based on an example and the example of a comparison, this invention is not limited to the following examples.

[0030] an example 1 -- first, it mixed so that the ratio (mole ratio) of CsBr (the Wako Pure Chem make, trade name: cesium bromide purity 99.9% 035-12062) and CsI (the Wako Pure Chem make, trade name: cesium iodide purity 99.9% 035-12442) might be set to 95:5, and mixed powder (sample) was obtained. Next, after putting in the above-mentioned mixed powder 6-8g into the cylinder container made from a quartz (20mmφ×35mmL), the inside of a container was made into the vacua (degree of vacuum: 10-5Torr), and the container was sealed. Then, the above-mentioned container has been arranged on the elevated-temperature band of a vertical mold furnace with a predetermined temperature gradient (elevated-temperature band: 670 degrees C, low-temperature band: 420 degree C), and the above-mentioned mixed powder was changed into the melt condition. Then, the above-mentioned container was slowly dropped on the low-temperature band from the above-mentioned elevated-temperature band so that a temperature fall rate might serve as 1.2-4.5 degrees C/hr, and the above-mentioned melt was made to crystallize from the lower limit.

[0031] The presentation of the obtained crystal was CsBr 0.95I0.05, and the consistency was 4.44 g/cm³. The crystal was cut to 10mmφ×2mmT, and the sample scintillator of this invention was obtained.

[0032] (Measurement of the damping time) The damping time of the above-mentioned scintillator was measured with the delayed coincidence counting method (delayed-coincidence method) using the measuring device shown in drawing 3. In the equipment shown in drawing 3, BaF₂ scintillator 50 is used as a reference and it connects with the

start input of the time amount-amplitude conversion circuit (TAC) 53 through the photo-multiplier (PMT, Hamamatsu Photonics make) 51 and the constant fraction discriminator (CFD) 52. Moreover, ^{22}Na is used as a radiation source 54, and it is arranged so that BaF2 scintillator 50 and the above-mentioned sample scintillator 55 may counter at equal intervals on both sides of a radiation source 54. Furthermore, PMT (Hamamatsu Photonics make) 57 enclosed by the lead block 56 is arranged so that the radiation from a radiation source 54 may not carry out direct incidence and the scintillation light from the sample scintillator 55 may serve as a single photon, and it connects with the stop input of TAC 53 through a preamp (fast amp) 58, CFD 59, and a delay circuit 60.

[0033] And the following Measuring-condition: radiations: Gamma ray (511 keV) While asking for the attenuation data of the fluorescence emitted from the sample scintillator 55 under ** with the multi-channel analyzer (MCA) 61 and approximating attenuation data using the least square method by computer 62, it asked for the exponential function of the number according to the number of fluorescence components as mentioned above. The attenuation data, decay curve, and exponential function which were obtained are shown in drawing 4.

[0034] For 1.4 ns, the damping time (τ_{short}) about an early fluorescence component was 13.5 ns, and was [the damping time (τ_{long}) about a late fluorescence component of the ratio ($S_{\text{short}}/S_{\text{long}}$) of the quantity of light of an early fluorescence component to the quantity of light of a late fluorescence component] 0.22 so that clearly from the exponential function shown in drawing 4. In addition, although the fluorescence component of 140 ps (es) was also contained, since the damping time (τ_{short}) was over the resolution (~ 200 ps) of a gaging system, it did not consider as the candidate for examination here.

[0035] (Measurement of relative luminescence reinforcement) The spectrum to each wavelength of the fluorescence emitted from the sample scintillator under following Measuring condition: temperature: 300 K was measured as an X-ray and a spectroscopy as the radiation source, using Hamamatsu Photonics 955 [trade name: R-] as trade name: M4QIII made from Carl Zeiss, and PMT. When were compared with the luminescence reinforcement of NaI (Tl) which calculated similarly the integral value (luminescence reinforcement) of the fluorescence spectrum, and the latter luminescence reinforcement was made into 100%, the former relative luminescence reinforcement (luminous efficiency) was 17.5%.

(Measurement of the temperature dependence of a fluorescence spectrum, and the temperature dependence of luminescence reinforcement) The temperature dependence of the fluorescence spectrum emitted from the sample scintillator, using Hamamatsu Photonics 955 [trade name: R-] as trade name: M4QIII made from Carl Zeiss and PMT was measured as an X-ray and a spectroscopy as the radiation source. The obtained result is shown in drawing 5. Moreover, the temperature dependence of luminescence reinforcement is searched for from the measurement data, and the result is shown in drawing 6.

[0036] Except having mixed so that the ratio (mole ratio) of example 2 CsBr and CsI might be set to 90:10, the crystal was obtained like the example 1 and the sample scintillator of this invention was obtained using the crystal. The presentation of the obtained crystal was CsBr 0.90I0.10, and the consistency was 4.44 g/cm³.

[0037] Using the above-mentioned sample scintillator, it asked for the attenuation data of

fluorescence like the example 1, and asked for the exponential function of the number according to the number of fluorescence components based on the attenuation data. The attenuation data, decay curve, and exponential function which were obtained are shown in drawing 7 . For 0.72ns, the damping time (τ_{short}) about an early fluorescence component was 10.7ns, and was [the damping time (τ_{long}) about a late fluorescence component of the ratio ($S_{\text{short}}/S_{\text{long}}$) of the quantity of light of an early fluorescence component to the quantity of light of a late fluorescence component] 0.15 so that clearly from the exponential function shown in drawing 7 .

[0038] Moreover, the spectrum to each wavelength of fluorescence was measured like the example 1 using the above-mentioned sample scintillator. The obtained result is shown in drawing 8 . Moreover, when were asked for relative luminescence reinforcement from the measurement data, and luminescence reinforcement of NaI (Tl) was made into 100%, relative luminescence reinforcement (luminous efficiency) was 13.9%.

[0039] Furthermore, the temperature dependence of a fluorescence spectrum was measured like the example 1 using the above-mentioned sample scintillator. The obtained result is shown in drawing 9 . Moreover, the temperature dependence of luminescence reinforcement is searched for from the measurement data, and the result is shown in drawing 10 .

[0040] Except having mixed so that the ratio (mole ratio) of example 3CsBr and CsI might be set to 99:1, the crystal was obtained like the example 1 and the sample scintillator of this invention was obtained using the crystal. The presentation of the obtained crystal was CsBr 0.99I0.01, and the consistency was 4.44 g/cm³.

[0041] When the spectrum to each wavelength of fluorescence was measured like the example 1, and it asked for relative luminescence reinforcement from the measurement data using the above-mentioned sample scintillator, and luminescence reinforcement of NaI (Tl) was made into 100%, relative luminescence reinforcement (luminous efficiency) was 17.1%.

[0042] Furthermore, the temperature dependence of a fluorescence spectrum was measured like the example 1 using the above-mentioned sample scintillator. The obtained result is shown in drawing 11 . Moreover, the temperature dependence of luminescence reinforcement is searched for from the measurement data, and the result is shown in drawing 12 .

[0043] Except having mixed so that the ratio (mole ratio) of example of comparison 1CsBr and CsI might be set to 10:90, the crystal was obtained like the example 1 and the sample scintillator was obtained using the crystal. The presentation of the obtained crystal was CsBr 0.10I0.90, and the consistency was 4.5 g/cm³.

[0044] When the spectrum to each wavelength of fluorescence was measured like the example 1, and it asked for relative luminescence reinforcement from the measurement data using the above-mentioned sample scintillator, and luminescence reinforcement of NaI (Tl) was made into 100%, relative luminescence reinforcement (luminous efficiency) was 5.1%.

[0045] Furthermore, the temperature dependence of a fluorescence spectrum was measured like the example 1 using the above-mentioned sample scintillator. The obtained result is shown in drawing 13 . Moreover, the temperature dependence of luminescence reinforcement is searched for from the measurement data, and the result is shown in drawing 14 .

[0046] Except having used only example of comparison 2CsBr as a raw material, the crystal was obtained like the example 1 and the sample scintillator was obtained using the crystal. The presentation of the obtained crystal was CsBr and the consistency was 4.44 g/cm³.

[0047] When the spectrum to each wavelength of fluorescence was measured like the example 1, and it asked for relative luminescence reinforcement from the measurement data using the above-mentioned sample scintillator, and luminescence reinforcement of NaI (Tl) was made into 100%, relative luminescence reinforcement (luminous efficiency) was 0%.

[0048] The scintillator of this invention emits the fluorescence which contains the fluorescence component with the very short damping time of 2 or less ns in a room temperature, and, moreover, its luminous efficiency was also fully excellent so that clearly from the result shown in drawing 4 and drawing 7 . On the other hand, the scintillator of this invention out of range had low luminous efficiency.

[0049] Moreover, the scintillator of this invention had high luminous efficiency at the room temperature, and the property that the damping time is short was acquired so that clearly from the result shown in drawing 5 , drawing 6 , drawing 9 , drawing 10 , drawing 11 , and drawing 12 .

[0050]

[Effect of the Invention] It becomes possible to emit the fluorescence which contains a fluorescence component with the very short damping time in a room temperature, maintaining luminous efficiency excellently according to the scintillator of this invention, as explained above. Moreover, since the fluorescence emitted by the scintillator of this invention contains a fluorescence component with the very short damping time in a room temperature as mentioned above, it becomes possible [obtaining a radiation detector with very high resolution] by using the component.

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

[Drawing 1] It is the graph which shows the attenuation data, decay curve, and exponential function for explaining the analysis approach of a decay curve.

[Drawing 2] It is the cross section showing 1 suitable operation gestalt of the radiation detector of this invention.

[Drawing 3] It is the block diagram showing the equipment configuration for measuring the damping time.

[Drawing 4] It is the graph which shows the attenuation data in the scintillator obtained in the example 1, a decay curve, and an exponential function.

[Drawing 5] It is the graph which shows the temperature dependence of the fluorescence spectrum in the scintillator obtained in the example 1.

[Drawing 6] It is the graph which shows the temperature dependence of the luminescence reinforcement in the scintillator obtained in the example 1.

[Drawing 7] It is the graph which shows the attenuation data in the scintillator obtained in the example 2, a decay curve, and an exponential function.

[Drawing 8] It is the graph which shows the fluorescence spectrum in the scintillator obtained in the example 2.

[Drawing 9] It is the graph which shows the temperature dependence of the fluorescence spectrum in the scintillator obtained in the example 2.

[Drawing 10] It is the graph which shows the temperature dependence of the luminescence reinforcement in the scintillator obtained in the example 2.

[Drawing 11] It is the graph which shows the temperature dependence of the fluorescence spectrum in the scintillator obtained in the example 3.

[Drawing 12] It is the graph which shows the temperature dependence of the luminescence reinforcement in the scintillator obtained in the example 3.

[Drawing 13] It is the graph which shows the temperature dependence of the fluorescence spectrum in the scintillator obtained in the example 1 of a comparison.

[Drawing 14] It is the graph which shows the temperature dependence of the luminescence reinforcement in the scintillator obtained in the example 1 of a comparison.

[Description of Notations]

10 [-- An antireflection film, 20 / -- The photomultiplier tube, 21 / -- A fluorescence input screen, 22 / -- A hermetic container, 23 / -- The photoelectric surface, 24 / -- A convergence electrode, 25 / -- A dynode, 26 / -- An anode, 30 / -- The radiation source, 31 / -- A radiation, 32 / -- Fluorescence, 33 / -- Photoelectron.] -- A scintillator, a 11 -- tubed container, 12 -- A fluorescence output screen, 13 -- A radiation input screen, 14

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(54) 【発明の名称】 シンチレータ及びそれを用いた放射線検出器

(57) 【要約】

【課題】 発光効率が高くかつ室温において極めて減衰時間が短い蛍光成分を含む蛍光を発するシンチレータ、並びに分解能の高い放射線検出器、を提供すること。

【解決手段】 放射線の入射に応じて所定の減衰時間を有する蛍光成分を含む蛍光を発するシンチレータであって、該シンチレータが、下記一般式：

$$\text{CsBr}_{1-x}\text{I}_x$$

【式中、xは0.01～0.1の数である。】で表される組成を有する結晶からなり、かつ前記蛍光成分の減

(2)

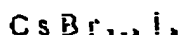
特開2000-249768

1

【特許請求の範囲】

【請求項1】 放射線の入射に応じて所定の減衰時間を有する蛍光成分を含む蛍光を発するシンチレータであって、

該シンチレータが、下記一般式：



【式中、 x は0.01～0.1の数である。】で表される組成を有する結晶からなり、かつ、前記蛍光成分の減衰時間が室温にて2 ns以下である。ことを特徴とするシンチレータ。

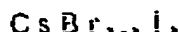
【請求項2】 前記蛍光が、所定の減衰時間を有する蛍光成分を複数含んでおり、該蛍光成分のうちの少なくとも一つの成分の減衰時間が室温にて2 ns以下であることを特徴とする。請求項1に記載のシンチレータ。

【請求項3】 前記結晶が、下記一般式：



【式中、 x は0.01～0.1の数である。】で表される組成となるように混合されたCsBrとCsIとの混合粉末を垂直ブリッジマン法によって結晶化せしめたものであることを特徴とする。請求項1又は2に記載のシンチレータ。

【請求項4】 放射線の入射に応じて所定の減衰時間を有する蛍光成分を含む蛍光を発するシンチレータであって、該シンチレータが、下記一般式：



【式中、 x は0.01～0.1の数である。】で表される組成を有する結晶からなり、かつ、前記蛍光成分の減衰時間が室温にて2 ns以下であるシンチレータと、前記シンチレータから発せられた蛍光を検出する蛍光検出器と、を備えることを特徴とする放射線検出器。

【発明の詳細な説明】

【0001】

【発明の属する技術分野】本発明は、放射線の入射に応じて蛍光を発するシンチレータ、並びにそれを用いた放射線検出器に関する。

【0002】

【従来の技術】従来、無機シンチレータの中で、減衰時間(decay time、蛍光寿命ともいう)が比較的短く、計測速度の向上が可能なものとして、CsI(ヨウ化セシウム)シンチレータが高エネルギー粒子の検出や医療機器

2

(カリウム)、Br(臭素)、Cl(塩素)等を不純物としてドーブした結晶からなるシンチレータが検討されており、CsIに少量のBrをドーブすることによって減衰速度が向上することが開示されている(A.Gektin, N.Shiran, V.Shlyahurov, A.Belsky "Development of Fast Scintillators on the Basis of CsI Doped with Homologous Impurities" Proc. Int. Conf. on Inorganic Scintillators and Their Applications, SCINT95 (1995) Delft University Press, The Netherlands, p. 415-418)。しかしながら、CsIに少量のBrをドーブした場合であっても減衰時間の減少はごく僅かであり、未だ充分なものではなく、一方、Br濃度が高くなると結晶が不安定となって分解が発生すると従来は認識されていた。

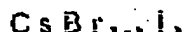
【0004】

【発明が解決しようとする課題】本発明は、上記従来技術の有する課題に鑑みてなされたものであり、発光効率が高くかつ室温において極めて減衰時間が短い蛍光成分を含む蛍光を発するシンチレータ、並びに分解能の高い放射線検出器を提供することを目的としている。

【0005】

【課題を解決するための手段】本発明者らは、上記目的を達成すべく鋭意研究を重ねた結果、CsBrに少量のIをドーブした場合に、発光効率が高くかつ極めて減衰時間が短い蛍光成分を含む蛍光を発するシンチレータを得られる可能性があることを見出し、本発明を完成するに至った。

【0006】すなわち、本発明のシンチレータは、放射線の入射に応じて所定の減衰時間を有する蛍光成分を含む蛍光を発するシンチレータであって、このシンチレータが、下記一般式：



【式中、 x は0.01～0.1の数である。】で表される組成を有する結晶からなり、かつ、前記蛍光成分の減衰時間が室温にて2 ns以下である。ことを特徴とするものである。

【0007】また、本発明の放射線検出器は、(a)放射線の入射に応じて所定の減衰時間を有する蛍光成分を含む蛍光を発するシンチレータであって、このシンチレータが、下記一般式：

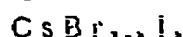
(3)

特開2000-249768

3

【0009】先ず、本発明のシンチレータについて説明する。

【0010】本発明のシンチレータは、下記一般式：



で表される組成を有する結晶からなるものである。上式中の x は、結晶中のBr（臭素）原子とI（ヨウ素）原子との合計数（モル数）に対するI原子の数の比率（モル分率）であり、原料としてCsBr（臭化セシウム）とCsI（ヨウ化セシウム）との混合物を用いる場合は原料中のCsIのモル分率に相当する。

【0011】本発明においては、上記 x が0.01以上0.1以下の範囲の数であることが必要である。 x が0.01未満では蛍光強度が弱くなり、他方、 x が0.1を超えると混晶ができず、Iが析出し、格子欠陥が増加する。また、上記 x は0.05以上0.08以下の範囲の数であることが好ましい。 x が上記下限未満ではIの数が減少し、蛍光強度が弱くなる傾向にあり、他方、 x が上記上限を超えるとIの析出による格子欠陥が増加し、早い減衰時間の成分が減少する傾向にある。

【0012】加えて、上記本発明のシンチレータは、放射線の入射に応じて、室温における減衰時間が2 ns以下、好ましくは1.5 ns以下、である蛍光成分を含む蛍光を発するものである。このように室温における減衰時間が2 ns以下という、極めて減衰時間が短い蛍光成分を含む蛍光が上記組成の結晶から発せられるということは、従来は全く認識されておらず、本発明者によって初めて見出された事実にほかならない。そして、本発明のシンチレータによって発せられる蛍光は上記のように室温において極めて減衰時間が短い蛍光成分を含むため、その成分を用いることによって非常に分解能の高い放射線検出器が得られる。他方、減衰時間が短いほどそのシンチレータを用いた放射線検出器の分解能の向上が可能となるが、減衰時間が100 ps未満ではシンチレータと組み合わせて用いられる蛍光検出器の分解能を超えてしまう傾向にあることから、上記減衰時間は100 ps以上であることが好ましい。なお、ここでいう「室温」とは、300 K (27℃)をいう。

【0013】また、本発明のシンチレータによって発せられる蛍光は、その発光メカニズムに基づいて通常複数の減衰成分（蛍光成分）を含んでおり、その場合はそれ

4

ず、蛍光が一つの成分からなる場合は、その減衰曲線は以下の指数関数：

$$F(t) = k \cdot e^{-t/\tau}$$

【式中、 t は時間、 $F(t)$ は時間に対する蛍光強度の関数、 k は蛍光強度の初期値（ $k = F(0)$ ）、 τ は減衰時間、をそれぞれ示す】で表される。そして、減衰時間は蛍光強度が初期値の $1/e$ に減衰するまでの時間であり、以下の関係：

$$F(\tau)/F(0) = 1/e$$

10 を満たす値である。

【0015】また、蛍光が二つの成分からなる場合は、図1に示すように、その減衰曲線は二つの指数関数の和、すなわち以下の関数：

$$\begin{aligned} F(t) &= f_1(t) + f_2(t) \\ &= k_1 \cdot e^{-t/\tau_1} + k_2 \cdot e^{-t/\tau_2} \end{aligned}$$

【式中、 t は時間、 $F(t)$ は時間に対する蛍光強度の関数、 $f_1(t)$ は第1成分についての時間に対する蛍光強度の関数、 $f_2(t)$ は第2成分についての時間に対する蛍光強度の関数、 k_1 は第1成分についての蛍光強度の初期値（ $k_1 = f_1(0)$ ）、 k_2 は第2成分についての蛍光強度の初期値（ $k_2 = f_2(0)$ ）、 τ_1 は第1成分についての減衰時間、 τ_2 は第2成分についての減衰時間、をそれぞれ示す】で表される。そして、第1及び第2成分についての減衰時間はそれぞれ、第1及び第2成分についての蛍光強度が初期値の $1/e$ に減衰するまでの時間であり、以下の関係：

$$f_1(\tau_1)/f_1(0) = 1/e$$

$$f_2(\tau_2)/f_2(0) = 1/e$$

を満たす値である。

30 【0016】更に、蛍光が三つ以上の成分からなる場合も同様に、その減衰曲線はその成分数に応じた指数関数の和で表され、各成分についての減衰時間は各成分についての蛍光強度が初期値の $1/e$ に減衰するまでの時間として求められる。

【0017】従って、計測データ（蛍光の減衰データ）を最小二乗法を用いて近似すると共に、その成分数に応じた数の指数関数を求め、得られた指数関数に基づいて各成分についての減衰時間が求められる。そして、蛍光が複数の成分からなる場合は、得られた減衰時間がより短い成分がより早い成分に対応する。なお、このような

(4)

特開2000-249768

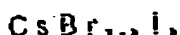
5

て変化するが、概ね287nmである。

【0019】更に、本発明のシンチレータの密度は概ね4.44g/cm³と比較的高く、それゆえにγ線等の放射線に対するストップパワー、ひいては吸収係数が高い。また、本発明のシンチレータは、NaI(Tl)の発光強度を100%とした時の相対発光強度(発光効率)が13.9~17.5%であり、従来のCsIシンチレータよりも発光効率が高い。

【0020】上記本発明のシンチレータを製造する方法は特に制限されないが、大型結晶の合成が可能ないわゆる垂直ブリッジマン法が好ましい。垂直ブリッジマン法とは、試料の融液を入れた縦長のろつばを所定の温度勾配をもつ縦型炉(結晶成長炉)の中でゆっくり降下させ、ろつば中の融液の下端から固化させて結晶を得る方法である。具体的には、以下の方法が特に好ましい。

【0021】(i) 先ず、下記一般式：



[式中、xは0.01~0.1の数である。]で表される組成となるようにCsBrとCsIとを混合して混合粉末(試料)を得る。

【0022】(ii) 次に、上記混合粉末を縦長のろつば(例えば石英容器)の中に入れた後、ろつば内を真空状態(真空度：10⁻³~10⁻¹Torr)にしてろつばを密封する。

【0023】(iii) その後、上記ろつばを、所定の温度勾配(高温帯：640~670℃、低温帯：420~450℃)をもつ縦型炉の高温帯に配置して、上記混合粉末を融液状態にする。

【0024】(iv) 続いて、上記ろつばを、降温速度が1.2~4.6℃/hrとなるように上記高温帯から低温帯にゆっくり降下させ、上記融液をその下端から結晶化せしめる。それによって、上記本発明のシンチレータを構成する結晶が効率良くかつ確実に得られる。

【0025】次に、本発明の放射線検出器について説明する。図2に、本発明の放射線検出器の好適な一実施形態を示す。同図に示す放射線検出器は、上記本発明のシンチレータ10と、蛍光検出器としての光電子増倍管20とを備えている。具体的には、筒状容器11の中にシンチレータ10が配置されており、筒状容器11の蛍光出方面12以外の放射線入方面13には反射防止膜14

5

成されて蛍光32が発せられる。次いで、蛍光出力面12及び蛍光入方面21を通過した蛍光によって光電子増倍管20内の電子が励起されて真空中に光電子33が放出される。そして、光電子33は収束電極24によって収束され、ダイノード25によって増倍(二次電子増倍)された後、二次電子群(電荷)がアノード26から取り出される。

【0027】このようにして、放射線のエネルギーに比例した量(強度)の蛍光がシンチレータ10において発生し、その蛍光量(蛍光強度)に比例した電荷が光電子増倍管20によって出力されるため、結果的には光電子増倍管20の出力パルス波高が放射線のエネルギーに比例する。従って、出力パルスの波高値と計数率を測定することによって放射線のエネルギー分布とその量を測定することができる。加えて、本発明のシンチレータによって発せられる蛍光は、前述のように室温において2ns以下という極めて減衰時間が短い蛍光成分を含むため、その成分を用いることによって放射線検出器の分解能の向上が可能となる。

【0028】以上、本発明の放射線検出器の好適な実施形態について説明したが、本発明の放射線検出器は上記実施形態に限定されるものではない。例えば、上記実施形態では蛍光検出器として光電子増倍管を使用しているが、蛍光検出器はフォトダイオード等であってもよい。また、上記実施形態ではラインフォーカス型の光電子増倍管を使用しているが、それ以外の型(例えば、ボックス型、サーキュラージュ型、ベネシアンブライント型、ファインメッシュ型、マイクロチャンネルプレート型)であってもよい。

【0029】

【実施例】以下、実施例及び比較例に基づいて本発明をより具体的に説明するが、本発明は以下の実施例に限定されるものではない。

【0030】実施例1

先ず、CsBr(和光純薬(株)製、商品名：臭化セシウム純度99.9% 035-12052)とCsI(和光純薬(株)製、商品名：ヨウ化セシウム純度99.9% 035-12442)との比率(モル比)が95:5となるように混合して混合粉末(試料)を得た。次に、上記混合粉末6~8gを石英製円筒容器(20mmφ×35mmL)の中に入れた後、

(5)

特開2000-249768

7

ンチレータを得た。

【0032】(減衰時間の測定) 図3に示す測定装置を使って、遅延同時計数法(delayed-coincidence method)によって上記シンチレータの減衰時間を測定した。図3に示す装置においては、リファレンスとしてBaF₂シンチレータ50が用いられ、光電子増倍管(PMT、浜松ホトニクス(株)製)51及び定フラクショナルデスクリミネータ(CFD)52を介して時間-振幅変換回路(TAC)53のスタート入力に接続されている。また、放射線源54として²⁴Naが使用され、放射線源54を換えてBaF₂シンチレータ50と上記試料シンチレータ55とが等間隔で対向するように配置されている。更に、放射線源54からの放射線が直接入射せずかつ試料シンチレータ55からのシンチレーション光がシングルフォトンとなるように鉛ブロック56で囲われたPMT(浜松ホトニクス(株)製)57が配置され、前置増幅器(fast amp)58、CFD59及び遅延回路60を介してTAC53のストップ入力に接続されている。

【0033】そして、以下の測定条件：

放射線：γ線(511keV)

の下で試料シンチレータ55から発せられた蛍光の減衰データを多チャンネルアナライザー(MCA)61で求め、コンピュータ62により減衰データを最小二乗法を用いて近似すると共に、蛍光成分数に応じた数の指数関数を前述のようにして求めた。得られた減衰データ、減衰曲線及び指数関数を図4に示す。

【0034】図4に示した指数関数から明らかなように、早い蛍光成分についての減衰時間(τ_{early})が1.4ns、遅い蛍光成分についての減衰時間(τ_{late})が13.5nsであり、遅い蛍光成分の光量に対する早い蛍光成分の光量の比($S_{\text{early}}/S_{\text{late}}$)は0.22であった。なお、減衰時間(τ_{early})が140psの蛍光成分も含まれていたが、測定システムの分解能(〜200ps)を超えているのでここでは検討対象としなかった。

【0035】(相対発光強度の測定) 放射線源としてX線、分光器としてカール・ツァイス社製の商品名：M4 QIII、PMTとして浜松ホトニクス(株)製の商品名：R-955を用いて、以下の測定条件：
温度：300K

8

シンチレータから発せられた蛍光スペクトルの温度依存性を測定した。得られた結果を図5に示す。また、その測定データから発光強度の温度依存性を求め、その結果を図6に示す。

【0036】実施例2

CsBrとCsIとの比率(モル比)が90：10となるように混合した以外は実施例1と同様にして結晶を得、その結晶を用いて本発明の試料シンチレータを得た。得られた結晶の組成はCsBr_{0.9}I_{0.1}であり、その密度は4.44g/cm³であった。

【0037】上記試料シンチレータを用い、実施例1と同様にして蛍光の減衰データを求め、その減衰データに基づいて蛍光成分数に応じた数の指数関数を求めた。得られた減衰データ、減衰曲線及び指数関数を図7に示す。図7に示した指数関数から明らかなように、早い蛍光成分についての減衰時間(τ_{early})が0.72ns、遅い蛍光成分についての減衰時間(τ_{late})が10.7nsであり、遅い蛍光成分の光量に対する早い蛍光成分の光量の比($S_{\text{early}}/S_{\text{late}}$)は0.15であった。

【0038】また、上記試料シンチレータを用い、実施例1と同様にして蛍光の各波長に対するスペクトルを測定した。得られた結果を図8に示す。また、その測定データから相対発光強度を求めたところ、NaI(Tl)の発光強度を100%とした時に相対発光強度(発光効率)は13.9%であった。

【0039】更に、上記試料シンチレータを用い、実施例1と同様にして蛍光スペクトルの温度依存性を測定した。得られた結果を図9に示す。また、その測定データから発光強度の温度依存性を求め、その結果を図10に示す。

【0040】実施例3

CsBrとCsIとの比率(モル比)が99：1となるように混合した以外は実施例1と同様にして結晶を得、その結晶を用いて本発明の試料シンチレータを得た。得られた結晶の組成はCsBr_{0.99}I_{0.01}であり、その密度は4.44g/cm³であった。

【0041】上記試料シンチレータを用い、実施例1と同様にして蛍光の各波長に対するスペクトルを測定し、その測定データから相対発光強度を求めたところ Na

(5)

特開2000-249768

9

10

得、その結晶を用いて試料シンチレータを得た。得られた結晶の組成は $\text{CsBr}_{0.9999}\text{I}_{0.0001}$ であり、その密度は 4.5 g/cm^3 であった。

【0044】上記試料シンチレータを用い、実施例1と同様にして蛍光の各波長に対するスペクトルを測定し、その測定データから相対発光強度を求めたところ、 NaI(Tl) の発光強度を100%とした時に相対発光強度(発光効率)は5.1%であった。

【0045】更に、上記試料シンチレータを用い、実施例1と同様にして蛍光スペクトルの温度依存性を測定した。得られた結果を図13に示す。また、その測定データから発光強度の温度依存性を求め、その結果を図14に示す。

【0046】比較例2

CsBr のみを原料として用いた以外は実施例1と同様にして結晶を得、その結晶を用いて試料シンチレータを得た。得られた結晶の組成は CsBr であり、その密度は 4.44 g/cm^3 であった。

【0047】上記試料シンチレータを用い、実施例1と同様にして蛍光の各波長に対するスペクトルを測定し、その測定データから相対発光強度を求めたところ、 NaI(Tl) の発光強度を100%とした時に相対発光強度(発光効率)は0%であった。

【0048】図4及び図7に示した結果から明らかなように、本発明のシンチレータは、室温において2 ns以下という極めて減衰時間が短い蛍光成分を含む蛍光を発するものであり、しかも発光効率も充分に高水準であった。他方、本発明の範囲外のシンチレータは発光効率が低いものであった。

【0049】また、図5、図6、図9、図10、図11及び図12に示した結果から明らかなように、本発明のシンチレータは室温で発光効率がよく、しかも減衰時間が短いという特性が得られた。

【0050】

【発明の効果】以上説明したように、本発明のシンチレータによれば、発光効率を高水準に維持しつつ、室温において極めて減衰時間が短い蛍光成分を含む蛍光を発することが可能となる。また、本発明のシンチレータによって発せられる蛍光は上記のように室温において極めて*

*減衰時間が短い蛍光成分を含むため、その成分を用いることによって非常に分解能が高い放射線検出器を得ることが可能となる。

【図面の簡単な説明】

【図1】減衰曲線の解析方法を説明するための、減衰データ、減衰曲線及び指数関数を示すグラフである。

【図2】本発明の放射線検出器の好適な一実施形態を示す断面模式図である。

【図3】減衰時間を測定するための装置構成を示すブロック図である。

【図4】実施例1で得られたシンチレータにおける減衰データ、減衰曲線及び指数関数を示すグラフである。

【図5】実施例1で得られたシンチレータにおける蛍光スペクトルの温度依存性を示すグラフである。

【図6】実施例1で得られたシンチレータにおける発光強度の温度依存性を示すグラフである。

【図7】実施例2で得られたシンチレータにおける減衰データ、減衰曲線及び指数関数を示すグラフである。

【図8】実施例2で得られたシンチレータにおける蛍光スペクトルを示すグラフである。

【図9】実施例2で得られたシンチレータにおける蛍光スペクトルの温度依存性を示すグラフである。

【図10】実施例2で得られたシンチレータにおける発光強度の温度依存性を示すグラフである。

【図11】実施例3で得られたシンチレータにおける蛍光スペクトルの温度依存性を示すグラフである。

【図12】実施例3で得られたシンチレータにおける発光強度の温度依存性を示すグラフである。

【図13】比較例1で得られたシンチレータにおける蛍光スペクトルの温度依存性を示すグラフである。

【図14】比較例1で得られたシンチレータにおける発光強度の温度依存性を示すグラフである。

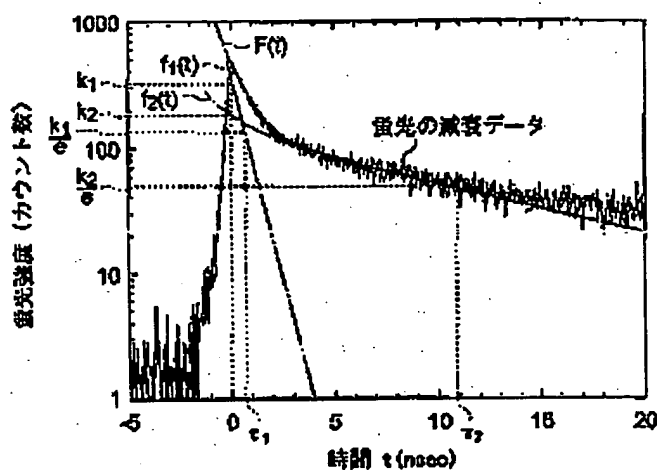
【符号の説明】

10…シンチレータ、11…筒状容器、12…蛍光出力面、13…放射線入力面、14…反射防止膜、20…光電子増倍管、21…蛍光入力面、22…密封容器、23…光電面、24…収束電極、25…ダイノード、26…アノード、30…放射線源、31…放射線、32…蛍光、33…光電子。

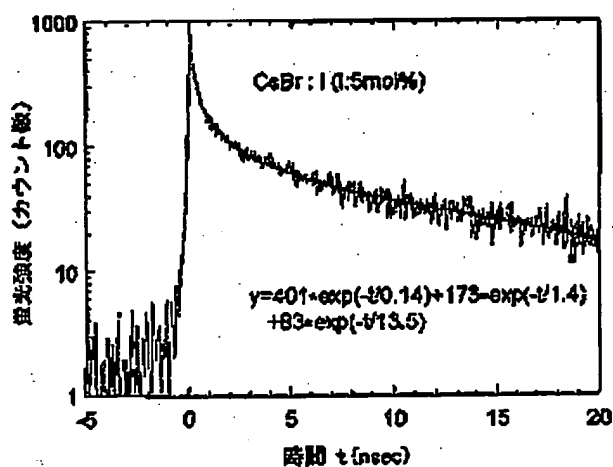
(7)

特開2000-249768

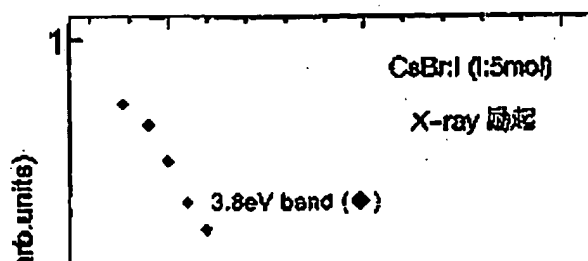
【図1】



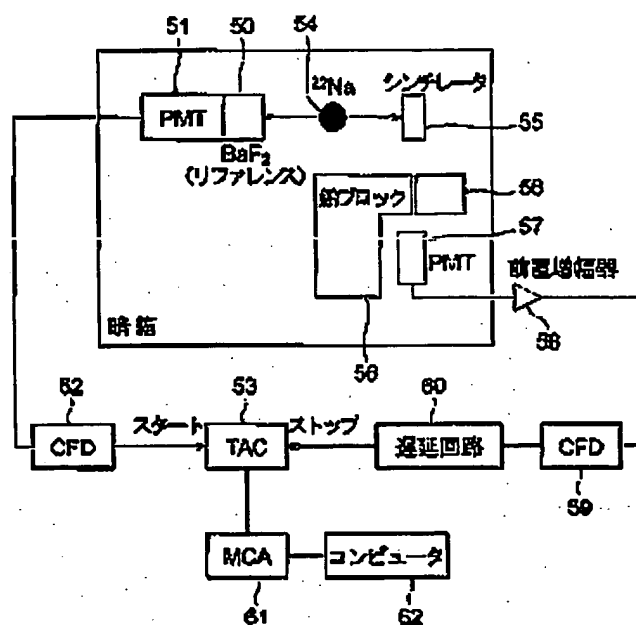
【図4】



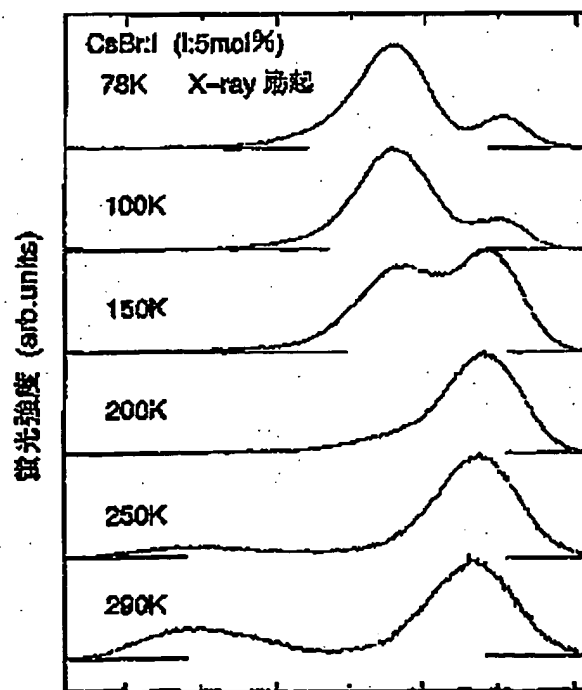
【図6】



【図3】



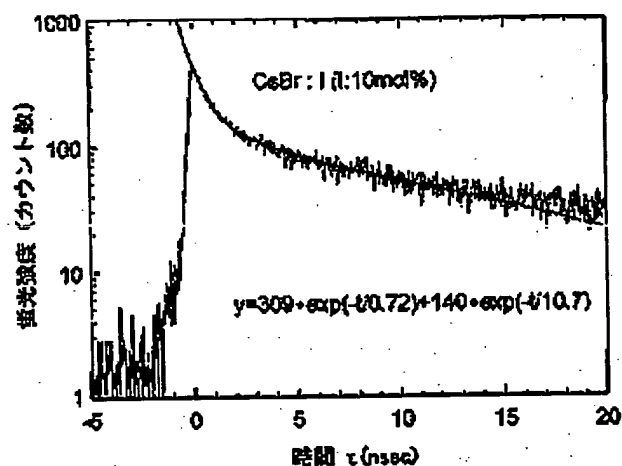
【図5】



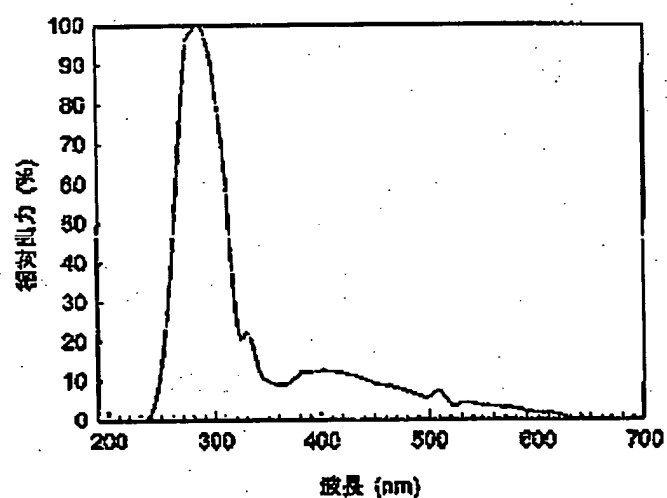
(8)

特開2000-249768

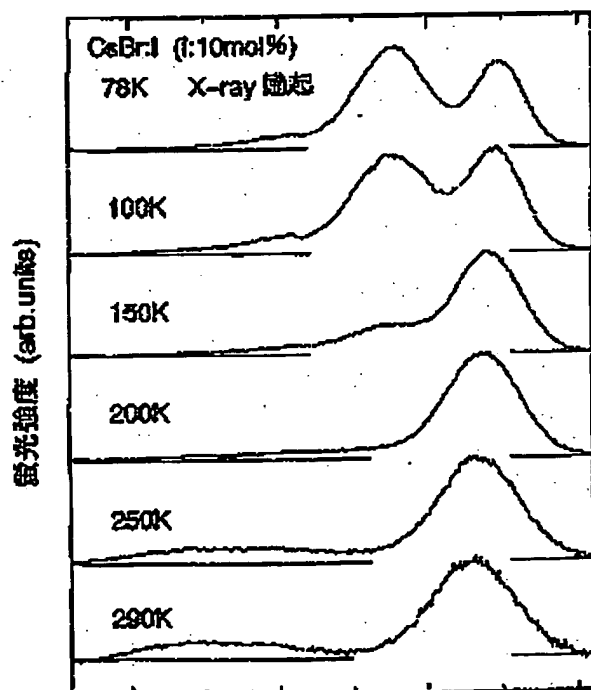
【図7】



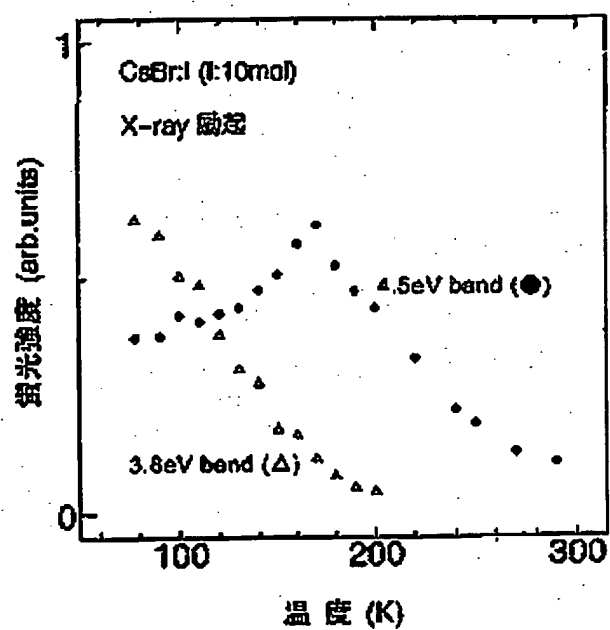
【図8】



【図9】



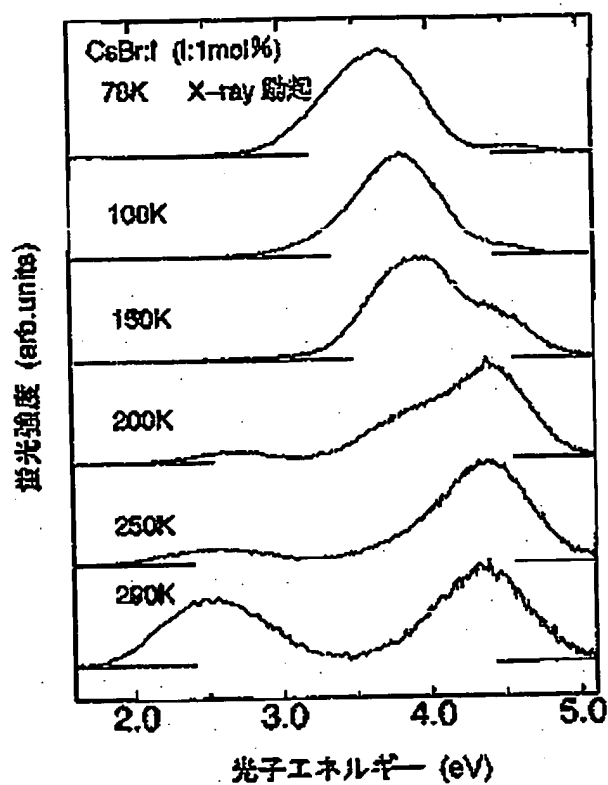
【図10】



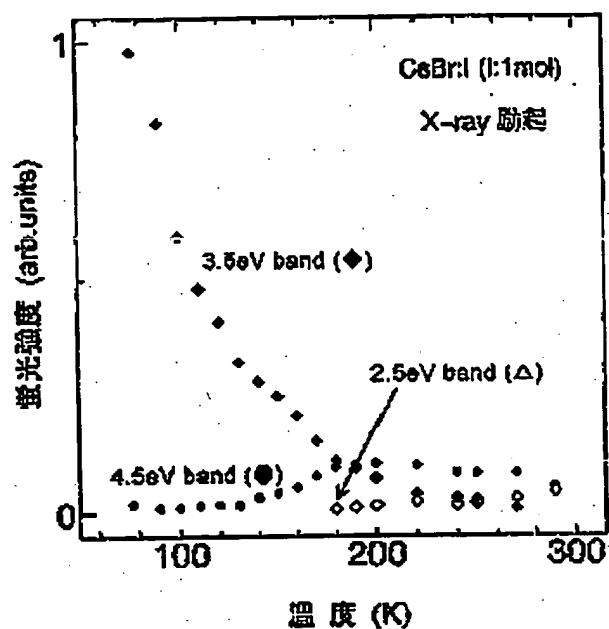
(9)

特開2000-249768

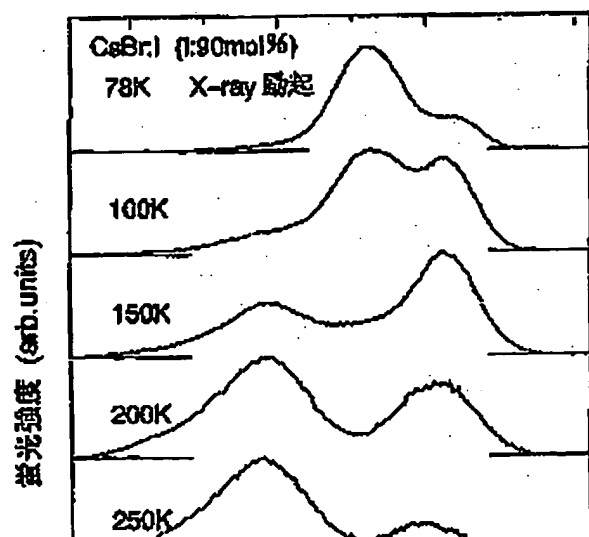
【図11】



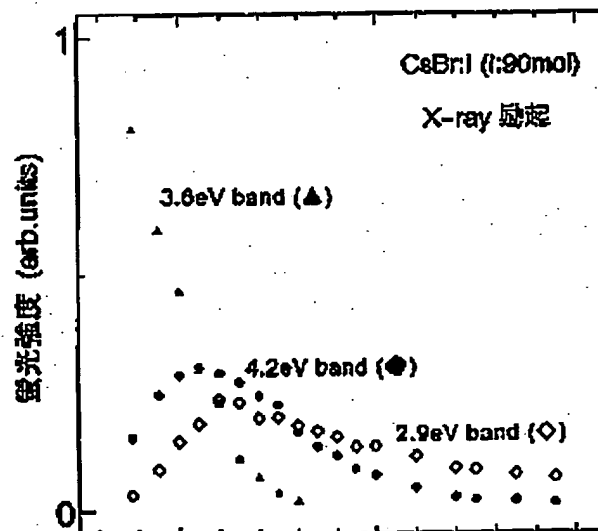
【図12】



【図13】



【図14】



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